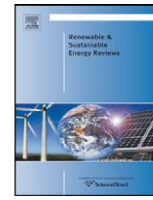
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Review of the membrane and bipolar plates materials for conventional and unitized regenerative fuel cells

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Fuel cell or hydrogen systems offer the potential for clean, reliable and on-site energy generation. This article review current literature with the objective of identifying the latest development in membrane and bipolar plates for the conventional fuel cell and unitized regenerative fuel cell (URFC). The result shows that the choice of both the bipolar plates and the catalysts for URFC electrodes is a delicate task, for bipolar plate the corrosion in the oxygen side will be the major problem and for the electrodes a very good candidate for fuel cell mode will not function well in the electrolyser mode and therefore it is suggested that a compromise should be considered. It is recommended that aluminum, titanium or for best results titanium with a gold-coated layer is a suitable candidate as the bipolar plate and Pt/IrO_X or Pt/Ru is suitable for an oxygen side catalyst in the URFC. For the conventional fuel cell the task is more easer because the corrosion problem is no more effective and thus the main goals for most of the studies was to concentrate on bipolar plate cost reduction, increase electrical conduction and reducing the platinum loading rate for catalyst.

Contents

1. Introduction

Fuel cell based power plants offer one of the most lucrative possibilities for future power generation and the fuel cell is also found to be potentially more efficient than the conventional plants since the fuel reacts electrochemically instead of combustion. In this way there is far less air, thermal, and noise pollution issues to be considered. A conventional fuel cell is an electrochemical device that produces electricity by separating the fuel (generally hydrogen gas) via a catalyst. The protons flow through a membrane and combine with oxygen to regenerate water with the help of the catalyst used while the electrons flow from the anode to the cathode to create electricity.

An electrolyser operates reversely to the fuel cell which splits water into hydrogen and oxygen by the power supplied. To make a hydrogen system self dependant energy source, an electrolyser should exist to generate gases which then be consume to generate power through the fuel cell. For some applications a specific weight (power per unit weight) is an important issue to be considered,

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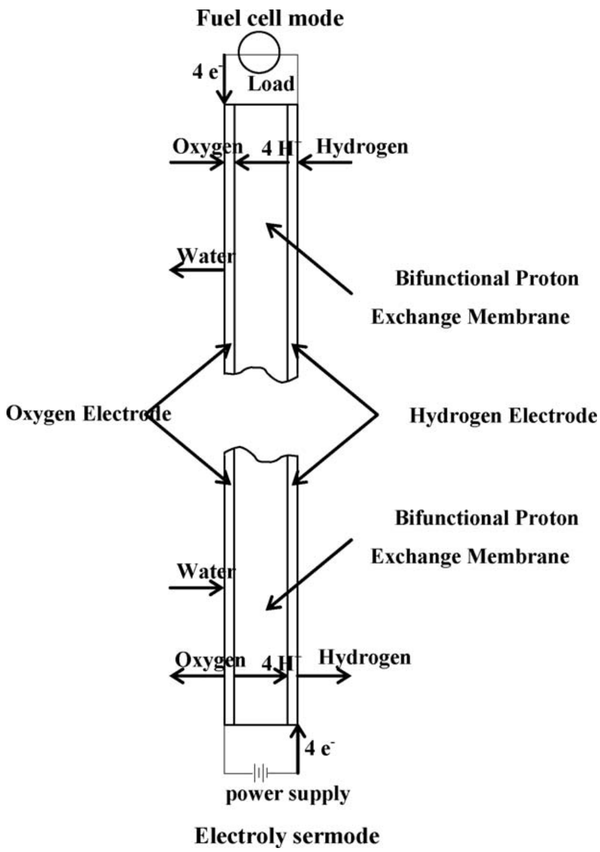


Fig. 1. Electrochemical reaction in the URFC.

thus researches succeed in getting one cell to function as electrolyser and fuel cell in one cell at different modes. This cell is known as a unitized regenerative fuel cell (URFC). The URFC works logically in the electrolyser mode first to store hydrogen and then use it in the fuel cell mode, thus the URFC behaves like a battery as a self independent power source but in contrast to the battery system, the URFC is unaffected by the depth of the discharge or length of the cycle duration. In the URFC, energy and power are not linked i.e. the cell is sized for power but the storage tank is sized for energy [1]. A diagram of the electrochemical reaction that occurs in the proton exchange membrane (PEM) of the URFC is shown in Fig. 1.

The objective of this study is to compare the materials used in a conventional fuel cell and the URFC. For that purpose a review of the latest development in the materials used for the bipolar plate and the development in the membrane electrode assembly (MEA) for both the conventional fuel cell and URFC was carried out.

2. Bipolar plate for conventional fuel cell

Flow channels are conveniently machined into a bipolar plate or field plate to allow a high electronic, good thermal conductivity and stability in a chemical environment inside a fuel cell [2]. In a fuel cell stack, each bipolar plate supports two adjacent cells and the bipolar plate is known to have five typical functions: (1) to distribute the fuel and its oxidants within the cell (2) to facilitate water management within the cell (3) to separate individual cells in the stack (4) to transport currents away from the cell and (5) to facilitate heat management [3]. The sketch of a bipolar plate is shown in Fig. 2.

The main target for all researchers is to focus on the bipolar cost reduction, increase the electric conductivity, decrease the weight and increase the corrosion resistance. Generally most of the bipolar plates used in conventional fuel cells are either made of graphite, stainless steel or of other metallic materials. In this article, we would like to review the latest development of each material individually.

2.1. Stainless steel

Stainless steel is widely used as a bipolar plate for fuel cell application that is due to its chemical stability during the chemical reaction and also its ability to conduct electric however, its corrosion resistivity is low unless treated or coated with some other material to enhance its corrosion resistivity. Hornung and Kappelt [4] examined the suitability of economical corrosion resistance of Fe-based alloys (FeBs) in the construction of bipolar plates. The results indicate that most of the (FeBs) exhibit a characteristic comparable to that of the Ni-based alloy. Makkus et al. [5] tested several stainless steel bipolar plates and found that using stainless steel as a flow plate at the anode side of a solid polymer fuel cell (SPFC) results in a higher contamination of the MEA as compared to a stainless steel flow plate that is used on the cathode side. Davies et al. [6] tested three stainless steel alloys

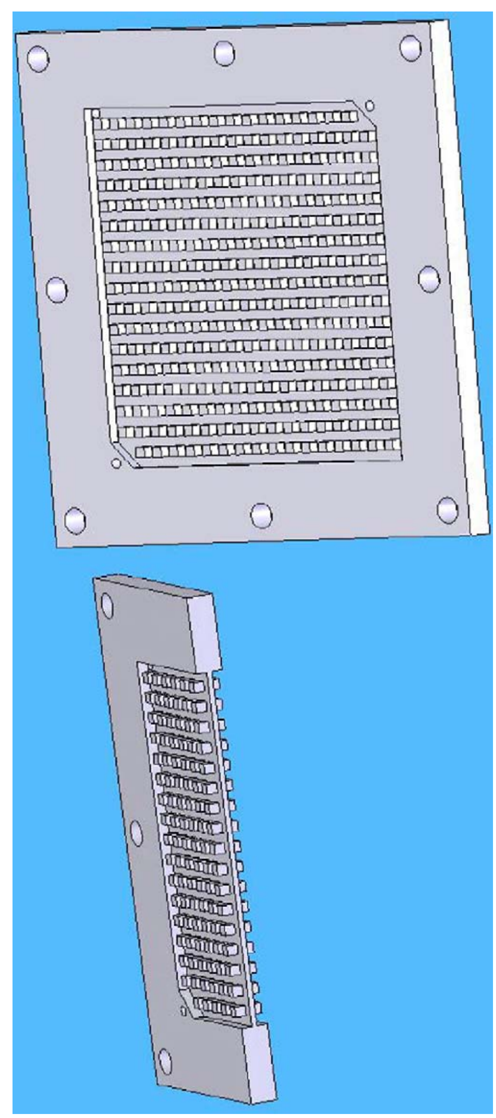


Fig. 2. Bipolar plate used in PEM fuel cell stack.

(310,316 and 904 L) as candidate bipolar materials for proton exchange membrane fuel cell (PEMFC). The fuel cell performance for the above bipolar materials is observed in the following order: 904 L < 310 < 316. Wang et al. [7] coated 316 L and 317 L stainless steel bipolar plates with 0.6 μm thick SnO_2:F. and this study suggests the possibility of using SnO_2:F coated with 317 L polymer electrolyte membrane fuel cell bipolar plate application.

Ren and Zeng [8] prepared compact titanium carbide as a coating for the type 304 stainless steel bipolar plate with a metallurgical bonding between the coating and the substrate. In this study it is found that TiC coating increases the corrosion potential and significantly decreases its corrosion current density. For the TiC-coated steel, it is observed to have no obvious degradation after a 30-day exposure in the solution. Lee et al. [9] coated thin stainless steel bipolar plates with 5 μm thick of multi-layered corrosion resistant material. The proposed manufacturing procedure proves to be a promising technology in producing metallic bipolar plates with micro-features.

Nam and Lee [10] evaluated the electrical and corrosion properties of surface-nitride AISI316L stainless steel bipolar plates for a PEMFC. The study shows that stainless steel with the Cr_2N nitride protective coating layer exhibits a better interfacial contact and corrosion resistance than the as-rolled or ($CrN + Cr_2N$)-coated AISI316L stainless steels.

Wang and Turner [11] tested Ferrite stainless steels (AISI441, AISI444, and AISI446) coated with 0.6 μm thick SnO_2:F bipolar plate simulated in PEMFC environment. The results show that a SnO_2:F coating enhances the corrosion resistance of the alloys and AISI446 shows an excellent corrosion resistance and the SnO_2:F coating seems to create additional resistance in the native air-formed films on these stainless steels. Wang and Northwood [12] used a Coated TiN on a martensitic stainless steel (SS410) as a bipolar plate. The results of this study show that this coating forms a much improved corrosion resistance on SS410 and therefore these coated materials could potentially be used in PEMFCs as a bipolar plate material provider. Lafront et al. [13] coated a stainless steel (316 L) bipolar plate with amorphous Zr75Ti25 alloy in simulated PEMFC in conditions of 25 °C and 80 °C. It is concluded from the corrosion data of this work that in the anode environment of a PEMFC, the $Zr_{75}Ti_{25}$ alloy could be a better candidate than 316 L for bipolar plates. The contrary is observed in the simulated cathode environment.

Jayaraj et al. [14] developed Fe- and Ni-base amorphous alloys as alternative bipolar plate materials for PEMFC. The results show that the Fe-base have lower interfacial contact resistance as compared to the Ni-base and both the Fe- and Ni-base amorphous alloys display a higher corrosion resistance than stainless steel. Wang and Northwood [15] coated SS316L with TiN as a developing metallic bipolar plate. The tests show that the corrosion resistance of SS316L significantly increases at 70 °C by coating with TiN for both anode and cathode conditions because the TiN-coated specimen is affected by pitting corrosion.

2.2. Graphite

One of the most well-established material for bipolar plate is graphite. Graphite is electrically conductive and reasonably easy to machine. It has also a very low density. However, it has three major disadvantages [16]:

- The machining of the graphite may be done automatically but the cutting takes time even on an expensive machine.
- Graphite is a brittle material and so the resulting cells need careful handling and the assembling is made difficult.
- Graphite is quite porous and therefore the plates need to be a few mm thicker to keep the reactant gases apart; this means that although the material has a low density, the final bipolar plate is not particularly light.

Due to these reasons above, many researchers work to enhance the performance of graphite bipolar plate such as, Scholta et al. [17] which presented a novel low cost graphite composite material bipolar plate as a promising candidate for PEMFC bipolar plate. Kakati and Deka [18] prepared composite bipolar plates for PEMFC by the compression molding technique using polymer as a binder and graphite as an electric filler material with some other reinforcements. Shen et al. [19] used sodium silicate/graphite composite as an acid corrosion resistant bipolar plate. The metal ionic content leach from this new bipolar plate is less than that from austenitic stainless steel. Dweiri and Sahari [20] adopted polypropylene, carbon black with graphite as prospective replacements for the traditional graphite bipolar plate in PEMFC. Yin et al. [21] mixed phenol formaldehyde resin powder with graphite powder as raw materials and used it as a kind of conductive composite for bipolar plate after a hot pressure molding process. The results show that: the conductivity decreases with the increase of PF resin content and the best conductivity is 142 s cm^{-1} when its PF resin content is 15% molded at 240 °C for 60 min. Radhakrishnan et al. [22] prepared hybrid composites bipolar plate consisting of high-temperature thermoplastic, graphite and a third additional conduction component.

Some others used other materials, Kumar and Reddy [23] used a porous material for a gas flow-field bipolar/end plate. It is observed, that the performance of fuel cell with Ni–Cr metal foam was highest, and decreased in the order SS-316 metal foam, conventional multi-parallel flow-field channel design and carbon cloth. Show [24] tested a Ti bipolar plate coated with amorphous carbon (a-C) film for PEMFC. The results show the fuel cell output is 1.4 times higher than the output of a bare (not a-C coated) Ti bipolar plate fuel cell. Maheshwari et al. [25] developed a polymer composite bipolar plate by making use of carbon fiber network in a specific form as a filler component. The plate when used in the unit fuel cell assembly shows *I–V* performance comparable to that of the commercially available bipolar plates but with a higher strength and stiffness.

3. Bipolar plate for (URFC)

A cell potential of 1.6–2 V in an electrolysis mode with O_2 is highly reactive and leads to a corrosive atmosphere. Thus, the bipolar plate material for URFC has to be corrosion resistant. Wittstadt et al. [26] choose titanium as a bipolar plate and end plate material for his URFC. Hodgson et al. [27] used coated titanium as a lightweight bipolar plate and it is found from this study that titanium can be employed to produce fuel cell with a very high volumetric and gravimetric power densities. Wind et al. [28] investigated the effect of coating on a metallic bipolar plate for PEM to prevent the formation of oxide layers with high receptivity. Hung et al. [29] compared the fuel cell performance of aluminium-coated bipolar plate with a conventional fuel cell bipolar plate made of graphite. The results indicate that coated metal bipolar plates show a 22% savings in hydrogen consumption, higher efficiency and durability and without any sign of power degradation.

The electric conductivity for titanium (refer to Table 1) is not as high as some other material but its high resistivity to corrosion makes it a very suitable candidate for URFC. For that reason an attempt to increase the electric conductivity was done by coating the surface of the titanium with a thin layer of gold. Wang et al. [30]

Table 1
Electrical conductivity for selected material.

Material	Electrical conductivity (S/m)
Gold	45.2×10^6
Aluminum	37.7×10^6
Ruthenium	3.19×10^6
Titanium	2.34×10^6
SS 316	1.33×10^6
Graphite	1.27×10^5

developed a light weight and corrosion resistant bipolar plate for the PEMFC. The bipolar plate was made from titanium and coated with gold. Wang et al. [31] investigated the effect of surface modification on the titanium bipolar plate. Two different surface modification materials, iridium oxide (IrO_2) and platinum were studied and the performance of these two plates is found to be very similar to the graphite plate.

4. Membrane electrode assembly (MEA) for URFC

Two problems are encountered when designing electrode structures for URFC systems. For PEM fuel cells, highly hydrophobitised carbon paper or carbon cloth is usually adopted as the gas diffusion layer or electrode materials but however, they cannot be used as the gas diffusion layer (GDL) of a URFC for the following two reasons: firstly, the carbon materials tend to corrode at high potentials on the oxygen electrode side during the water electrolysis operation; secondly, GDLs have to achieve an appropriate balance between the hydrophobic and hydrophilic properties for both the fuel cell and water electrolysis operations. The fuel cell operation requires that the oxygen GDL have a hydrophobic property to prevent water flooding, on the other hand, water electrolysis requires that the GDL has a hydrophilic property to supply water to the oxygen electrode [32]. A schematic sketch of a MEA for a fuel cell is shown in Fig. 3.

Some researches have demonstrated to overcome or reduce the effect of those two problems. Swette et al. [33] used $Na_xPt_3O_4$ as a bifunctional oxygen electrode catalyst in the URFC. The results show a highly efficient bifunctional oxygen electrocatalysis. This study also used another approach by integrating two catalysts into dual-character electrodes like Pt/IrO_2, Pt/RhO_2 or Rh/RhO_2.The same approach can be applied to a single bifunctional catalyst as such electrodes also show efficiency as electrodes. Ahn [34] developed a new concept of the bifunctional electrode. In this new concept, oxidation and reduction reactions are assigned to the electrodes. Thus hydrogen oxidation (fuel cell mode) and oxygen evolution (electrolysis mode) proceed alternatively on one electrode, namely the oxidation electrode, whereas the two remaining reactions proceed at the reduction electrode. The suggested cell operating with these types of electrodes is known as an 'integrated water electrolysis and hydrogen/oxygen fuel' as opposed to the 'reversible' fuel cell. By using this concept, it is found to be less difficult to identify suitable electrode material for both electrodes.

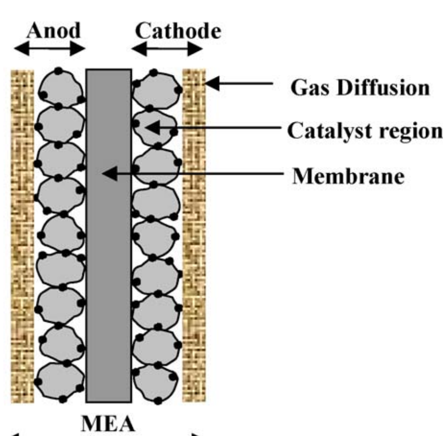


Fig. 3. A Schematic membrane electrode assembly.

Hari [35] tested a URFC with Pt: C ratio 20:80 at a loading of 1 mg/cm^2 as an electrode for the hydrogen side and Pt/Ir-on-C with a 20% iridium content and impregnated with 5% Nafion at approximately 0.5 mg/cm^2 as an electrode for the oxygen side. The data collection for the regenerative is first from the fuel cell mode and then the electrolyser shows better electrolysis performance and finally as it slightly lowers for Pt/Ir-on-C than those for Pt-on-C electrodes. Swette et al. [36] tested different oxygen electrode catalyst materials and different decompositions and compared the results with the baseline electrode with Pt at both the hydrogen and oxygen side. The results show that the regenerative PEM fuel cells display an efficient bifunctional performance that can be fabricated with the traditional Pt for the hydrogen side and Pt-Ir catalyst if the electrode structures are properly designed. This investigation has also identified RuO_x as a catalyst with a good potential for improving the O_2-evolution component of a bifunctional positive electrode. Ledjeff et al. [37] shows the manufacturing procedure of coating Nafion117 membrane with a variety of different noble catalysts such as Ru and Ir. The resulting electrodes are very thin and show a high performance.

Zhigang et al. [38] used platinum as a bifunctional catalyst for a hydrogen electrode and 50 wt. % Pt + 50 wt. % IrO_2 for a bifunctional catalyst of the oxygen electrode. The study also shows that using a thin catalyst layer can reduce the loading of the catalyst to 0.4 mg/cm^2 and minimizes mass transport and ohmic overvoltage at the same time giving the cell a promising stability under any number cycle operations. Prakash [39] investigated the effect of using the ruthenium as a catalyst for both oxygen reduction and generation in URFC. The results show that ruthenium is quite an effective catalyst for the oxygen reduction and generation reaction. However; it suffers stability problems in an anodic mode. The study proposes the use of anion-conducting polymers both as membrane over layers and as integral components of gas-diffusion electrode as it promises to alleviate this problem.

Ioroi [40] showed that the preparation procedure of the IrO_2/Pt catalyst can affect the performance of the URFC, comparing the result of catalyst prepared by mixing IrO_2/Pt and the one with deposited IrO_2/Pt. The deposited method shows 2–3% improvement of the roundtrip efficiency and this enhancement is due likely to the microstructure of the electrocatalyst. Chen et al. [41] prepared electrode arrays containing 715 unique combinations of five elements (Pt, Ru, Os, Ir, and Rh) by the Borohydride reaction of aqueous metal salts. Catalysts that showed high activity for both reactions and good resistance to anodic corrosion have been identified in the Pt–Ru rich region of the Pt–Ru–Ir ternary. The ternary catalyst $Pt_{4.5}Ru4Ir_{0.5}$ (subscripts indicate atomic ratios) is significantly more active than the previously described Pt_1Ir_1 bifunctional catalyst for both reactions.

Ioroi et al. [42] showed that the PTFE content in the electrocatalyst layer affected only the fuel cell performance. The electrode containing 5–7 wt. % PTFE was appropriate for URFC and also the results show that the Nafion content in the electrode affected both the fuel cell and water electrolysis performance; the electrode containing 7–9 wt. % Nafion showed good performance, and the addition of a small amount of iridium catalyst (about 10 at.

%) to the oxygen electrode layer significantly improved the URFC performance. Ioroi et al. [43] studied the relation between the polytetrafluroethylene (PTFE) coating amount on the gas diffusion backing (GDB) and the URFC performance. The PTFE coating on the GDB of the H_2 electrode does not affect the cell performance. But however, the URFC performance depends significantly on the coating of O_2 electrode side. The study also concludes that the cell with the smaller PTFE loading amount on the O_2 GDB exhibited a higher fuel cell and lower water electrolysis voltage. The results of the alternate operations between fuel cell and water electrolysis cycle tests reveal that a stable operation of the URFC cycle is achieved using the appropriate amount of PTFE loading on the GDB ($16\ mg\ cm^{-2}$).

Burke [44] also reported the testing of the URFC system for the fuel cell technology program at the Glenn Research Center (GRC) and plans to minimize the system weight, volume, and parasitic power as its goal. The design concept currently being developed uses no pumps to circulate coolants or reactants, and minimizes the ancillary components to only the oxygen and hydrogen gas storage tanks, a water storage tank, a loop heat pipe to control the temperature and two pressure control devices that control the cell stack pressure during operation.

Yim et al. [45] employed various oxygen electrode catalysts for URFC. The results show that Pt + Ir was the best in URFC's performance and cyclic stability, revealing the least activity loss in fuel cell mode and significant activity improvement in water electrolysis mode by adding Ir to Pt black. Burke [46] developed a unique lightweight URFC and the cell is unique in that it uses regenerative gas dryers/humidifiers that are mounted on the surface of the gas storage tanks that act as the radiators for thermal control of the URFC system. The system's function is to dry the hydrogen and oxygen gases produced by electrolysis when the gas storage tanks cool down during the charging mode and to humidify the hydrogen and oxygen gases used by the fuel cell.

Lee et al. [47] proposed a novel method of direct deposition of Pt catalysts on Nafion membrane modified with polypyrrole for the URFC. The study demonstrated that the polypyrrole could play a role as an electronic sink material for Pt catalyst deposition and the subsequent chemical reduction. Yim et al. [48] tested experimentally a single cell URFC system. The results show that the fuel cell mode performance in the order of Pt black > PtIr > $PtRuO_x$ > P-PtRu ~ PtRuIr > $PtIrO_x$ and water electrolysis performance PtIr ~ P-$PtIrO_x$ > PtRu > PtRuIr $\gtrsim$ Pt black. For both modes PtIr shows the best URFC performance and with increasing Ir or IrO_x composition from 0 to 50% in $PtIrO_x$ catalyst the fuel cell performance decreases while the water electrolysis significantly improves. Wittstadt et al. [26] sputtered platinum on the oxygen electrode for URFC after hot pressing. The main enhancement is caused by the higher fuel cell mode efficiency, the round-trip efficiency is 4% higher than the one of the standard MEA. Song et al. [49] developed a novel bifunctional oxygen electrode with a thin-film electrocatalyst layer and a corrosion-resistive gas diffusion layer (GDL) prepared by the carbon paper backing and a protective micro-porous layer (MPL). The protective MPL is made of the IrO_2 and deposits fine Ti powders. This electrode shows slight performance loss during 20 cycles.

Yao [50] prepared a bifunctional electrocatalyst for oxygen electrodes of URFC by chemical deposition of platinum on IrO_2 powder in aqueous solutions. The study also explains the differences between the deposition and mixing methods of preparing Pt/IrO_2 on the electrode surface. The results show that the oxygen electrode with IrO_2-supported Pt prepared by the deposition method exhibits slightly lower oxygen reduction reaction (ORR) activity but markedly a higher oxygen evolution reaction (OER) activity than a mixture of Pt and IrO_2. Zhang et al. [51] tested the performance of fuel cell/water electrolysis of URFC. The electrocatalyst for the oxygen side of the URFC is bifunctional RuO_2–IrO_2/Pt .The results show that there are differences in morphology and structure between deposited RuO_2–IrO_2/Pt and mixed RuO_2–IrO_2/Pt. and that the performance of URFC using deposited RuO_2–IrO_2/Pt electrocatalyst with a high dispersion is better than that of URFC using mixed RuO_2–IrO_2/Pt electrocatalyst.

5. Membrane electrode assembly (MEA) for conventional fuel cell

For the conventional fuel cell the problem of corrosion is less than the URFC because the cell works only as a fuel cell. However, continuous research has been carried out to enhance the ability of the membrane to produce the required chemical reaction. Towne et al. [52] developed new method for fabricating MEAs for PEMFCs using a home-made inkjet printer device to deposit successive layers of Pt/C catalyst. The cell successfully operated 10% less of catalyst loading than the commercial membrane which operates by 33% less of the catalyst loading of $0.2\ mg/cm^2$ of Pt approximately.

Taylor et al. [53] also used the inkjet printer device as a catalyst application method for PEMFCs and the results show that this method will lead to a very low platinum loading but the low loading is not easily attained when using conventional loading method. Gruber et al. [54] fabricated a mini fuel cell with maximum power density of $149\ mW/cm^2$ at a very low loading of $0.054\ mg/cm^2$ for both anode and cathode. Gruber et al. [55] also used sputter-deposited method for applying the Pt as a catalyst material for PEM fuel cells with an ultra low loading of $5\ \mu g/cm^2$. The author also studied the effect of using the sputter-deposited method on different porous electrodes and found an 8% performance improvement after adding chromium or by coating with a thin film-like layer of palladium. Moreira et al. [56] used Palladium (Pd) as a catalyst with Carbon and Vulcan as supporting materials. The results indicate that Pd, with Vulcan presented a better performance.

6. Conclusions

This paper reports the development on bipolar and membrane material for conventional and unitized regenerative fuel cells and the following conclusions from this study can be summarized as

1. Selecting titanium for URFC is a good choice in terms of its high resisitivity to corrosion but since its electric conductivity is not high so it is recommended to coat the titanium bipolar plate with a gold layer to enhance the conductance ability.
2. Choosing URFC oxygen side catalyst is a delicate task because it is a very good candidate for fuel cell mode and will be not suitable for the electrolyser mode, thus a compromise should be made to get an acceptable dual cell function by adopting IrO_X, Ru or both with platinum. This will be a very good candidate for the oxygen side URFC.
3. For the hydrogen side the mater is less difficult because the corrosion environment does exist any more in the hydrogen side and thus a Pt/C will be an acceptable candidate.
4. Most of the research on fuel cell component materials focus on cost and weight decrease and increase in the electrical conductivity, so therefore currently a metallic material has received more attention for it to be used as fuel cell bipolar plate selection material.
5. For the conventional fuel cell catalyst material most of the research focus on how to decrease the rate of platinum loading. The cost of membrane with an ultra low rating any how reaches less than $0.05\ mg/cm^2$ which is therefore considered a good performance.

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